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1    **Low black carbon concentration in agricultural soils of central and northern Ethiopia**

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15    **Abstract.** Soil carbon (C) represents the largest terrestrial carbon stock and is key for soil  
16    productivity. Major fractions of soil C consist of organic C, carbonates and black C. The turnover  
17    rate of black C is lower than that of organic C, and black C abundance decreases the vulnerability  
18    of soil C stock to decomposition under climate change. The aim of this study was to determine the  
19    distribution of soil C in different pools and impact of agricultural management on the abundance of  
20    different species. Soil C fractions were quantified in the topsoils (0–15 cm) of 23 sites in the  
21    tropical highlands of Ethiopia. The sites in central Ethiopia represented paired plots of agroforestry  
22    and adjacent control plots where cereal crops were traditionally grown in clayey soils. In the sandy  
23    loam and loam soils of northern Ethiopia, the pairs represented restrained grazing with adjacent  
24    control plots with free grazing, and terracing with cereal-based cropping with adjacent control plots  
25    without terracing. Soil C contained in carbonates, organic matter and black C along with total C was

determined. The total C median was 1.5% (range 0.3–3.6%). The median proportion of organic C was 85% (range 53–94%), 6% (0–41%) for carbonate C and 6% (4–21%) for black C. An increase was observed in the organic C and black C fractions attributable to agroforestry and restrained grazing. The very low concentration of the relatively stable black C fraction and the dominance of organic C in these Ethiopian soils suggest vulnerability to degradation and the necessity for cultivation practices maintaining the C stock.

Key words: black carbon, biochar, soil organic matter, carbonates, Ethiopia

## **Introduction**

Soil carbon (C) is usually divided into fractions contained in 1) soil organic matter (SOM), 2) carbonates and 3) various forms of black C. Soil organic C (SOC) contained in SOM has been estimated for large geographical areas (e.g., Batjes, 2002; Bradley et al., 2005; Heikkinen et al., 2013; Jones et al., 2005; Lugato et al., 2014) and even at a global scale (Batjes, 1996). Inorganic C (carbonates) estimates have also been published (e.g., Batjes, 1996; Rawlins et al., 2011; Shi et al., 2012). Black C is not routinely included in the estimates of soil C stocks, which globally consist of approximately 2400 Pg organic C in the top 200 cm and ca. 700 Pg of inorganic C in the top 100 cm (Batjes, 1996). However, black C represents a more persistent fraction of soil C stock in comparison with SOM while also black C may be affected by management.

SOM responds to land-use practices and climate change, and its decline is recognized as one of the eight soil threats in the EU Thematic Strategy for Soil Protection (EC, 2006). According to several studies in various countries (Bellamy et al., 2008; Guo & Gifford, 2002; Heikkinen et al., 2013), SOC has decreased in agricultural systems all over the world. Inorganic C in carbonates, in turn, declines upon soil acidification. By origin, black C is part of SOC, but is not considered a

51 part of SOM for two reasons: 1) results of the Walkley-Black wet digestion method, which many  
52 soil maps on SOC and SOM are still based on, does not include this fraction (Batjes, 1996) and 2)  
53 this C form is stable in soil, even for thousands of years (Atkinson et al., 2010; Lehmann et al.,  
54 2006). In this paper we chose to exclude black C from SOC, which is here defined as C contained  
55 in SOM.

56  
57 Black C is formed by pyrolysis. It mainly consists of 1) charcoal, 2) biochar and 3) soot (Preston &  
58 Schmidt, 2006). Charcoal occurs in soil predominantly as a consequence of forest fires, while  
59 deliberately added soil amendment is called biochar. From the chemical viewpoint, soot is similar to  
60 biochar and charcoal, even though it may contain more inorganic substances (DeLuca & Aplet,  
61 2008). Black C contributes to cation exchange capacity and water-holding capacity. Application of  
62 biochar to soil commonly increases yields (Jeffery et al., 2011, Laghari et al., 2016), however not in  
63 SOM-rich soils (Tammeorg et al., 2014). Kuhlbusch & Crutzen (1995) estimated that 50–250 Tg of  
64 charcoal annually enters soil and water ecosystems. Taking into account the stability of this C form,  
65 the black C stock in soil is probably large. Even though individual studies have been conducted on  
66 the concentration or stock of black C in soil (reviewed by Preston & Schmidt, 2006; Zhan et al.,  
67 2013), more measured data are needed to form accurate estimates equal to the other fractions of soil  
68 C.

69  
70 Because of the assumed stability of black C, ecological research did not consider it a relevant  
71 topic of investigation until a decade ago (DeLuca & Aplet, 2008). Interest was augmented by the  
72 *Terra preta* cultures of indigenous peoples of South American Indians (Glaser et al., 2000). The  
73 potential of this stabile soil C fraction to counteract and resist rapid climate change has brought it  
74 to the focus of attention.

75

76 Concentration of C in Ethiopian agricultural soils is commonly at a moderate level at the very  
 77 least, compared to many other tropical countries. For example, Sillanpää (1982) measured an  
 78 average SOC content of 2.2% in 71 agricultural soils of Ethiopia, while the mean for 574 soil  
 79 samples from six other African countries was only 1.3%. This result may partly be connected to  
 80 the high clay content (45%) of Sillanpää's Ethiopian soils compared to the rest of the sampled  
 81 African soils (22%). Shifting cultivation is still a common agricultural practice throughout the  
 82 tropical world, and burning of the vegetation results in a substantial input of black C into the soil  
 83 (Kuhlbusch & Crutzen, 1995; Rumpel et al., 2006). The area of natural forests in Ethiopia has  
 84 declined from approximately 40% to less than 3% in 100 years, part of it being subjected to fire  
 85 (Berhaun, 2005). Wood and charcoal are extensively used as fuel throughout Africa, and the  
 86 remaining ash and charcoal pieces are often spread in fields or in the soil of kitchen middens.  
 87 Ethiopian soils may therefore be high in black C.  
 88

89 In this study we investigated the distribution of black C, SOC and carbonates in agricultural soils  
 90 in two areas in Ethiopia. Conventional cereal production was practiced in the sampled fields, and  
 91 agroforestry, terracing and restrained grazing were practiced as improved management regimes.  
 92 In our earlier study (Rimhanen et al., 2016) it was found that agroforestry and restrained grazing  
 93 increased total C in soil. Now the objective was to obtain quantitative information concerning the  
 94 fractions of C, particularly of black C to advance understanding of the persistence of soil C stocks.  
 95 We hypothesized that there is a substantial pool of black C in our experimental soils and the  
 96 concentration of black C remains unchanged and gains of soil C appear solely in the more labile  
 97 organic C pool.  
 98  
 99  
 100

## 101    **Material and Methods**

102    The soil material in our study originated from Sire (mean elevation 1970 m asl, mean precipitation  
103    868 mm, mean temperature 15–20 °C ) in the Oromia region of the Central Rift Valley, Ethiopia,  
104    and in Kobo (1590 m asl, 631 mm, 21–25 °C) in the Amhara region, northern Ethiopia (Fig. 1),  
105    both representing important food-producing areas. Field plots with soil conservation and adjacent  
106    plots with traditional practices were sampled. The soil conservation practices represented in our  
107    study were farmland terracing and areas with restrained grazing in Kobo and agroforestry around  
108    the homesteads in Sire. Traditional “highland temperate mixed” farming (Dixon et al., 2001) was  
109    represented by cereal and lentil cultivation adjacent to terracing and agroforestry plots, and by  
110    grazing land adjacent to areas with restrained grazing where domestic animals were excluded. The  
111    median (and range) duration for the improved management was 8.5 (6–20) years for agroforestry,  
112    13 (6–17) years for restrained grazing and 7 (5–10) years for terraced plots. The study areas,  
113    agricultural practices, sampling and general soil properties have been described in detail in our  
114    previous study (Rimhanen et al., 2016), where total C concentrations and stocks were investigated.  
115    Briefly, soil (0–15 cm) was sampled from seven restrained grazing sites, eight terraced and  
116    agroforestry sites and from adjacent control plots for each study site. Three independent replicates,  
117    consisting of ten sub-samples, were collected from each plot with an auger. The Kobo area was  
118    dominated by loam and sandy loam soils, with 14% clay and 58% sand contents on average, and as  
119    much as 12% of coarse fragments, expressed on the volumetric basis. In Sire, the soil texture ranged  
120    from clay to clay loam, the average clay and sand contents being 39% and 24%, respectively, with  
121    only 3% of coarse fragments. The average pH (1:2.5 soil : water) of both areas was 7.8, with all  
122    results falling within the range of 7.4–8.2. The bulk densities, needed for calculating the C stocks  
123    and determined with a core method, ranged from 0.97 to 1.50 kg dm<sup>-2</sup>, with a mean of 1.25 kg dm<sup>-3</sup>.

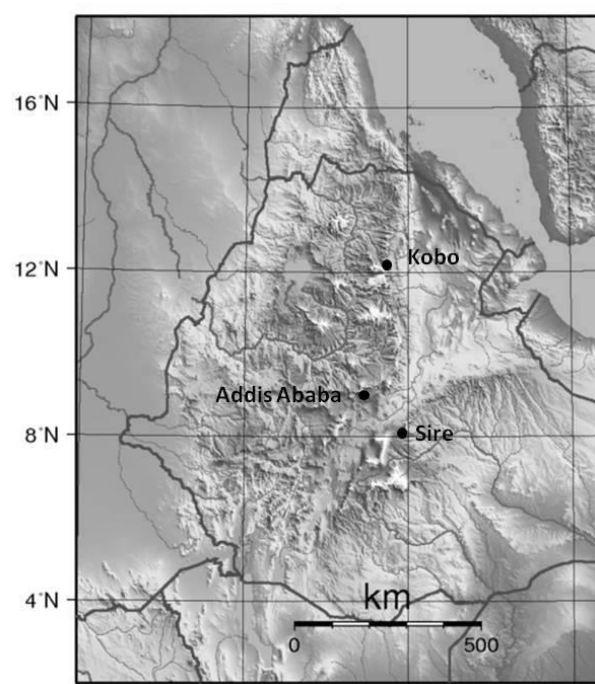


Fig. 1. Locations of the sampling areas of Sire and Kobo in central and northern Ethiopia.

### Determination of soil carbon

Total C of the fine earth fraction (< 2 mm) of all soil samples was analysed by dry combustion at 1100°C with a Leco CN-2000 analyser (Leco Corporation, MI, USA) or a VarioMAX CN- (Elementar Analysensysteme GmbH) using 500-mg soil samples. Carbon contained in the carbonates was determined indirectly by digesting 3-g soil samples with 5 ml of 6 M HCl for 10 min (Ellert & Rock, 2007). Thereafter the mixture was transferred on a filter paper (Whatman Ribbon 589/3) and washed five times with deionized water to avoid the possible corrosion of surfaces by acidic vapours during the further experimental steps. Dried soil samples (60 °C, 24 h) were analysed for total C, and the difference between untreated soil and HCl-treated soil was as the measure of carbonate C. Black C and SOC were determined with the fractionation method of Kurth et al. (2006), developed for measuring charcoal in forest soils. In this method, carbonaceous organic matter (SOM) are removed from the sample, and the total C remaining in the sample is assumed to be black carbon (C<sub>bc</sub>). Briefly, a 3-g soil sample was digested with 30 ml of 1 M HNO<sub>3</sub> and 60 ml of concentrated

140 at room temperature for 30 min. The mixture was boiled slowly for 16 hrs, divided into two  
141 consecutive days. The samples were dried (60 °C, 24 h), ground and analysed for total C, assumed  
142 to represent black C. Quartz sand was used as the blank. An estimate for SOC was obtained by  
143 subtracting the concentrations of carbonate C and black C from the total C of the untreated sample.  
144  
145 The fractionation method was tested in four soil samples (Table 1) from Finland by measuring the  
146 recovery of biochar added into the soil. The biochar was manufactured by Tammeorg et al. (2014)  
147 from spruce (*Picea abies*) that was pyrolyzed for 10–15 min at 550–600 °C. C content of the  
148 biochar material was 81.3%, and 5.5% of C was contained in carbonates. The biochar additions  
149 were 0, 4 and 8 g of biochar material into 400-g samples of air-dry soil (three replicates),  
150 corresponding to 0, 30 and 60 t ha<sup>-1</sup> in a 25-cm plough layer. The soil-biochar mixtures were  
151 incubated at a moisture of 20% (sand) or 25% (clay) for 30 days at 5 °C. Dried soil samples were  
152 analysed for the fractions of C as presented above in three replicates. Total C of the unamended soil  
153 samples ranged from 0.8% to 3.4%. While SOC clearly dominated in the topsoils, all test soil  
154 samples had a substantial concentration of black C (0.3–0.6%). Addition of biochar elevated total C  
155 (Fig. 2 for sand, results of clay not presented in detail). As a response to the carbonates contained in  
156 the biochar material, some increase in carbonate C fraction was observed in all soil samples but no  
157 change was observed in the SOC fraction. The most remarkable change after biochar addition was  
158 measured in the black C fraction. In the sandy soil, 90% of C contained in the added biochar was  
159 recovered during the fractionation but on average 63% was recovered from the clay soil. These  
160 results indicate that the fractionation method is able to quantify different C pools at least  
161 satisfactorily. The average coefficients of variation (CV), calculated based on the replicates, were  
162 5.2%, 9.0% and 12.8% for total C, SOC and black C, respectively, while the CV for the small  
163 carbonate fraction was 34%.

164

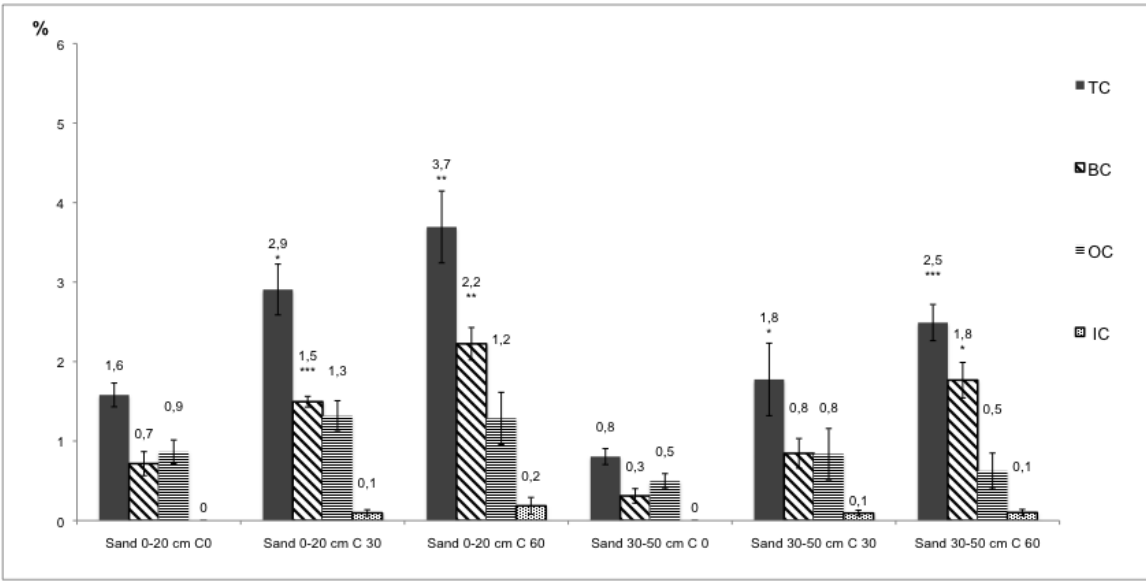


165 Table 1. Chracteristics of the soil samples used for testing the fractionation method for C.

Characteristic	Clay 0–20	Clay 30–50	Sand 0–20	Sand 30–50
	cm	cm	cm	cm
Texture	Sandy clay loam	Clay	Sand	Sand
Clay %	24	46	4	2
Silt %	34	39	16	5
Sand %	42	15	80	93
pH(H <sub>2</sub> O)	6.4	5.9	5.8	6.2

166

167



168

169 Fig. 2. Carbon (C) contents in the sandy topsoil (0–20 cm) and subsoil (30–50 cm) at three rates of  
170 added biochar: C 0 = 0 t ha<sup>-1</sup> (control), C 30 = 30 t ha<sup>-1</sup> and C 60 = 60 t ha<sup>-1</sup>. TC = total C, BC =  
171 charcoal/biochar, OC = C in organic matter, IC = C contained in carbonates. The standard  
172 deviations are presented as error bars. Statistical significance of the differences in TC, BC and IC  
173 compared to the control: \*\*\* p= 0.001, \*\* p= 0.01, \* p= 0.05.

174

175 **Data analysis.** The statistical analysis of the soil C concentrations (SOC, black C and their  
176 proportions of total C) were based on generalized linear mixed models for a split-plot design, where  
177 the three groups of plot pairs (agroforestry, areas of restrained grazing and farmland terracing) were  
178 the levels of the whole-plot factor and the two management practices (traditional and improved)  
179 were the levels of the sub-plot factor. The models included three fixed effects (main effects of the  
180 whole-plot factor and the sub-plot factor and their interaction) and two random effects (whole-plot  
181 error and sub-plot error). The means of ten subsamples were used as observations in the statistical  
182 analyses.

183

184 The charcoal concentrations were normally distributed, but the distributions of other dependent  
185 variables were skewed. Generalized linear mixed models with beta (SOC %) and gamma (SOC and  
186 black C %) distributions were used in the analysis to satisfy the assumptions of the models (Gbur et  
187 al., 2012). Black C was fitted by using the restricted maximum likelihood (REML) estimation  
188 method and others by applying the residual pseudo-likelihood with a subject-specific expansion  
189 (RSPL). Degrees of freedom were calculated using the Kenward-Roger method. The normality of  
190 residuals was checked using box plots (Tukey, 1977). The residuals were also plotted against the  
191 fitted values. These plots indicated that the assumptions of the models were adequate. Comparison  
192 of the means was performed using two-tailed t-tests. The analyses were performed using the  
193 GLIMMIX procedure in version 9.3 of the SAS/STAT software (Littell, 2006).

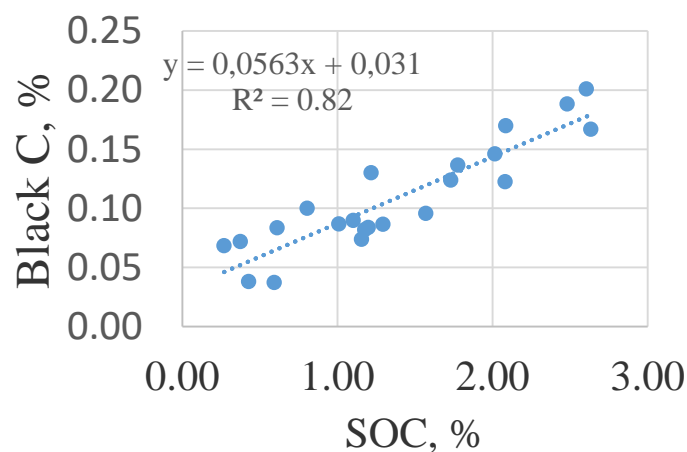
194

## 195 **Results**

196 The CVs, based on three independent replicates of each plot, indicated that the sampled plots were  
197 reasonably homogeneous, except for the small fraction of carbonates. For total C, SOC and black C,  
198 the average CVs were 10, 12 and 10%, respectively, but 49% for C in carbonates.

199

200 The clayey soils of Sire, central Ethiopia, contained more total C (mean 2.4%, median 2.2%) than  
 201 the drier and more coarse-textured soils of Kobo in northern Ethiopia (mean 1.0%, median 1.1%).  
 202 The terraced fields and their traditionally cultivated control plots in particular were lower in C  
 203 (mean 0.74%) than the adjacent plots with restrained grazing (mean 1.4%). The results of total C  
 204 and their relationship to cultivation practices have been presented and discussed in detail by  
 205 Rimhanen et al. (2016).  
 206  
 207 SOC was by far the dominating C fraction, with a median of 85% and a range of 53–94% of total C.  
 208 Despite the rather high pH, the soils were practically non-calcareous, with a carbonate C range of  
 209 0–0.86% and a mean of 0.08%. The mean corresponded to 0.7% and the maximum to 7% calcite,  
 210 while calcareous soils by definition contain at least 15% calcite. The carbonate C median was as  
 211 low as 0.07%, representing 6% of total C, with a range of 0–41%. Within the narrow range of  
 212 carbonate concentrations and soil pH (7.4–8.3), no correlation was observed between carbonate  
 213 content and soil pH. Although the highest concentrations occurred in soils with pH>8, soils of  
 214 similar pH with very low carbonate C concentrations were also observed.  
 215  
 216 Black C concentrations ranged from 0.03 to 0.24% of soil mass, with one outlier in Kobo that had a  
 217 concentration of 0.31%, while the concentration median was 0.10%. Only 6% of total C was  
 218 contained in black C, with a range of 4–21%. Black C comprised 10–15% of total C in four of the  
 219 terraces and their control plots that were very low in total C (0.4–0.9%) despite average-level  
 220 concentrations (ca. 0.07%) of black C, while all other results were 4–9% of total C. Despite the  
 221 narrow range of black C, Fig. 3 indicates that there was a close linear relationship between SOC and  
 222 black C concentrations.  
 223



224

225 Fig. 3. The relationship between the SOC, i.e., C contained in SOM, and black C concentrations in  
 226 the sampled plots in Ethiopia. SOC=soil organic carbon; SOM=soil organic matter; C=carbon

227

228 Among the soil conservation practices, the plots for restrained grazing and agroforestry had  
 229 statistically significantly higher concentrations of SOC and black C than the control plots (Table 2).  
 230 The concentrations of carbonate C were not affected by the treatments.

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241 Table 2. Test results for the comparison of the soil conservation (cons) and traditional (trad)  
 242 management practices in terms of SOC (C contained in soil organic matter) and black C. The  
 243 results are presented without one discrepant value for the traditional control in a pair of farmland  
 244 terracing. N=number of plots.

Management practice	N	SOC, %			Black C, %		
		Mean and 95% CI <sup>2)</sup> in trad	Difference (cons-trad) <sup>1)</sup>	P-value	Mean and 95% CI <sup>2)</sup>	Difference (cons-trad)	P-value
Agroforestry (Sire)	16	1.74 1.34–2.25	0.55	0.028*	0.14 0.12–0.16	0.04	0.013*
Restrained grazing (Kobo)	14	0.98 0.75–1.30	0.66	0.001***	0.07 0.05–0.10	0.04	0.019*
Terracing (Kobo)	15	0.49 0.38–0.64	0.13	0.066 <sup>ns</sup>	0.06 0.04–0.09	0.01	0.72 <sup>ns</sup>

245 1) The estimated means for improved and traditional management practices are presented in the  
 246 original scale and the difference between the resulting values was calculated. The differences were  
 247 tested on the link scale using two-tailed t-tests.

248 2) 95% confidence intervals (CIs) for the means

249

250 Using the fixed depth approach, the stocks of the different C pools were calculated for the top 15  
 251 cm of soil, corresponding to the sampling depth. The mean stock of black C in the loam and sandy

252 loam soils of Kobo was  $1.2 \text{ t ha}^{-1}$ . This stock was quite independent of the treatments (restrained  
253 grazing, terracing, traditional cereal cultivation) and, compared to the control, was only  $0.2 \text{ t ha}^{-1}$   
254 higher in the fields where restrained grazing was practised while C contained in SOM was increased  
255 from  $13 \text{ t ha}^{-1}$  by  $4.5 \text{ t ha}^{-1}$ . In the clay and clay loam soils of Sire the mean stock of black C in the  
256 traditionally cultivated soils was  $2.8 \text{ t/ha}$  and  $0.7 \text{ t ha}^{-1}$  higher in the agroforestry plots. The  
257 improved management mostly influenced the SOC pool which amounted to  $36.4 \text{ t ha}^{-1}$  in the control  
258 plots and was  $9.3 \text{ t ha}^{-1}$  higher in the agroforestry plots.

259

## 260 **Discussion**

261 Agroforestry and restrained grazing increase the input of plant residues into the soil and elevate  
262 the concentration of soil C (Rimhanen et al., 2016). The present results show that most of the  
263 increase by far took place in the SOC fraction but there was also a statistically significant increase  
264 in the fraction of supposedly black C. According to the interviews, no marked fires occurred in the  
265 studied areas during the improved practices (Rimhanen et al., 2016). There are two alternative  
266 causes for the measured increase in black C. First, ash mixed with charcoal may have been used as  
267 a fertilizer in the improved practices. Second, and more likely, because the outcome was uniform,  
268 part of the litter had been incorporated into forms that were too recalcitrant or not accessible for  
269 oxidization by the  $\text{HNO}_3\text{-H}_2\text{O}_2$  treatment. Kurth et al. (2006) mentioned that digestion effectively  
270 removed *most* organic C. This conclusion is also supported by the high linear correlation between  
271 the fractions of SOC and black C. Therefore C remaining in soil after the  $\text{HNO}_3\text{-H}_2\text{O}_2$  –digestion,  
272 besides black C, may contain SOC forms that are chemically most stable or physically protected.  
273 Even though this fraction may not be purely black C, it likely represents the soil C pool that is  
274 most resistant against oxidation and which may thus form a buffer against the decline of SOC.

275 Terracing contributes to decreased erosion and may thereby result in more C remaining in the  
276 field. Since terracing itself does not increase C inputs into the soil, no statistically significant  
277 changes in C fractions were observed.

278

279 Our results did not support the hypothesis that soils of northern and central Ethiopia are high in  
280 black C. On the contrary, on an average only 6% of soil C was found in the fraction resistant  
281 against the  $\text{HNO}_3$  -  $\text{H}_2\text{O}_2$  oxidation. This black C content is much lower than reported in studies of  
282 cooler climates. According to Kurth et al. (2006), black C in five agricultural topsoils of northern  
283 USA had a range 0.29–0.92%, representing an average 17% of total C and in another five US  
284 soils, black C represented 10–35% of total C (Skjemstad et al., 2002), and in Australia black C  
285 amounted up to 40% of total C (Skjemstad et al., 1996). However, our results of black C  
286 correspond to other areas of warm climates. In the <2-mm fraction of two sandy savannah soils in  
287 Zimbabwe (Bird et al. 1999), “oxidation-resistant elemental C (OREC)” stood for 3.6 and 2.2% of  
288 SOC, or 0.6 and 1.0% of soil dry weight. In six surface horizons of agricultural soils of Laos  
289 (Rumpel et al., 2006), OREC constituted 5.5–7.3% of SOC, or 1.2–2.7% of soil. In a large  
290 material of 260 soil samples from the Chinese loess plateau (Zhan et al., 2013), black C  
291 concentration averaged at 0.07%, and in agreement with our results, was higher in clayey soils  
292 than in coarse-textured soils.

293

## 294 **Conclusions**

295 Most C accumulated in the soil during the application of agroforestry and restrained grazing was  
296 contained in SOM oxidized by a  $\text{HNO}_3$  -  $\text{H}_2\text{O}_2$  treatment. Minor increases of more resistant forms  
297 of C were also measured. As most C in Ethiopian soils is contained in SOC, these soils are likely  
298 very susceptible to the adverse effects of organic matter decline. Therefore sustainable use of these

299 soils strongly calls upon practices that contribute to the maintenance and continuous build-up of  
300 soil organic matter.

301

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308 in demanding conditions. We thank Ms. Merja Eurola, M.Sc. and the staff at the MTT Soil  
309 laboratory for the analyses of the Sire and Kobo soils soils.

310

## 311 **Author Contributions**

312 K.R. and H.K. selected the management practices and study sites. K.R. collected the samples and  
313 organized the data. J.M. carried out the experiment on biochar recovery. H.K. supervised the study  
314 regarding the Sire and Kobo soils and M.Y.H the biochar recovery study. J.K. designed and  
315 performed the data analyses. All authors contributed to the study design and to writing the  
316 manuscript.

317

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